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B. Remarks Concerning the Amendment

Please enter an amendment to Claim 1 as shown in Appendix A attached hereto. Claim 1 is amended to incorporate the words ---with oxygen--- after the words "for use in an oxidation process" and before the words "in the presence of hydrogen". Support for this amendment is found at page 4, lines 23-25, of the specification.

Claim 1 has been amended to specify that the catalyst so prepared is "for use in an oxidation process with oxygen in the presence of hydrogen." The words "with oxygen" are added for clarification. The prior claim language "for use in an oxidation process in the presence of hydrogen" may have caused confusion. Hydrogen is not an oxidant, but rather a reductant. The present amendment clarifies that the oxidation is taking place with oxygen as the oxidant and that additionally hydrogen is present in the process. The words "with oxygen" are not intended to be limited to the use of molecular oxygen as the only source of oxygen; but rather are intended to include the full scope of oxygen sources, including air, mixtures of oxygen with inert gases, such as helium, and other equivalents. Moreover, the amended phrase "for use in an oxidation process with oxygen in the presence of hydrogen" has been incorporated into Claim 1 as one means to distinguish the claimed process from the prior art, as noted in Section C hereinafter.

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C. Remarks Concerning the Office Action

1. Concerning Allowable Subject Matter

The Applicants gratefully acknowledge the Examiner's finding that Claims 8, 10-17, and 19-21 would be allowable, if rewritten in independent form including all of the limitations of the base claim and any intervening claims.

2. Brief Review of the Invention and its Benefits

The claimed invention pertains to a process of preparing a catalyst composition comprising gold on a titanium-containing support. The process comprises impregnating a gold compound and a reducing agent onto the catalyst support, wherein the catalyst support comprises titanium, or the reducing agent comprises titanium, or both the catalyst support and the reducing agent comprise titanium. The catalyst thus prepared is to be employed in an oxidation process with oxygen in the presence of hydrogen.

As compared with various prior art aqueous precipitation methods of preparing gold-titanium catalysts, the claimed catalyst preparation beneficially requires less solvent; may employ non-aqueous solvent; does not require controlling pH; can be conducted relatively quickly; provides for improved control over the amount of gold deposited; wastes little gold; and does not require recovery of unused gold from solution. Accordingly, the claimed method is more cost effective and more adaptable to commercialization.

3. Concerning the Rejection of Claims 1-7, 9, 18, 22-27, and 29-33

Claims 1-7, 9, 18, 22-27, and 29-33 stand rejected under 35 U.S.C. 102(b) as anticipated by US 5,502,020 (Iwakura), for the reasons of record. Note that Claim 33 is canceled. This rejection, as it might apply to amended Claims 1-7, 9, 18, 22-27, and 29-32 is traversed for the following reasons.

The problem solved by Applicants comprises this: How can a catalyst that comprises gold on a titanium-containing support and that is active and selective in a hydro-oxidation process be prepared by an impregnation

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method? The question is relevant, because the prior art, namely EP-A1-0,709,306 (copy provided in previous application file) teaches at page 7, that Au-Ti catalysts for use in hydro-oxidation processes cannot be prepared by impregnation, that the impregnation technique produces an inactive hydro-oxidation catalyst.

Applicants' solution to the problem, as specified in Claim 1, involves *impregnating a gold compound and a reducing agent* onto a titanium-containing support. The titanium may be derived from the reducing agent, the support, or both. Reducing agents that do not contain titanium include, for example, alcohols, such as ethanol.

Iwakura teaches an impregnation method of preparing a silver catalyst containing tungsten, cesium and an amine complex on a support, which may consist of titania. Gold is taught only as a peripheral element among a long list of promoter elements to be used in combination with the silver, tungsten, cesium, and amine. For disclosure of a reducing agent, the Office Action relies on Iwakura at Column 6, lines 39-43:

"Any of the above-described compounds can be deposited on the porous carrier in various ways. It is most realistic for the silver compound to be used in the form of an aqueous solution with an amine. The aqueous solution may have alcohol or the like added."

The clear gist of Iwakura involves employing alcohol as a *solvent* as an aid for dissolving the silver compound and the amine to produce a *silver-containing* impregnation solution. The language in no manner leads the skilled artisan to use an alcohol as a reducing agent in preparing a catalyst having gold as the primary catalytically-active species.

The Office Action refers to Example 21 and Table 2; but these citations merely disclose titanium ethoxylate $[\text{Ti}(\text{OCH}_2\text{CH}_3)_4]$ as a source of titanium in preparing a catalyst comprising silver, cesium, tungsten, and titania, *but not gold*. Water is the solvent, not ethanol. The reference makes no explicit or inherent teaching that *if* any ethanol were to be liberated, it would function

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beneficially as a reducing agent, more preferably, in the presence of a gold compound.

In point of fact, Iwakura is a forest of detailed components and elements, absent any "markers" or "trail blazers" that would anticipate the claimed combination of impregnating a gold compound and a reducing agent onto a titanium-containing support for the specific function of preparing an active hydro-oxidation catalyst.

Claim 1 also requires Applicants' prepared catalyst to be active in an oxidation process with oxygen in the presence of hydrogen ("hydro-oxidation process"). The Examiner cites Iwakura's teaching of "air" as supporting disclosure of oxidation with oxygen in the presence of hydrogen. This argument must fail. Air comprises 20.946 percent oxygen and only 0.5 parts per million hydrogen (equivalent to 0.00005 percent hydrogen), by volume. (See *Handbook of Chemistry and Physics*, 57th edition, CRC Press, 1976-1977, p. F-21, copy enclosed.) The molar ratio of oxygen to hydrogen in air exceeds 400,000:1. The trace amounts of hydrogen present in air are inconsequential to Iwakura's oxidation chemistry and the vast majority of air oxidations ubiquitously taught in the art. Iwakura in its entirety relates to oxidations using essentially oxygen ("direct oxidation") with no materially-effective quantity of hydrogen. In contrast, the claims require hydrogen as a *material element*, that is, present in a quantity sufficient to affect measurably the outcome of the process. Along the lines of Applicants' specification at page 19, lines 1-3, a material amount of hydrogen is typically greater than about 0.01 percent of the feed and, preferably, greater than about 0.1 percent of the feed, which values correlate to greater than about 200, preferably greater than about 2,000 times the quantity of hydrogen present in air.

The Examiner is reminded of the undersigned's presentation made in Arlington on May 7, 2003, wherein the undersigned explained the differences between a direct oxidation process (essentially oxygen) and a hydro-oxidation process (oxygen in the presence of hydrogen). The outcome of direct oxidation with oxygen includes partial oxidation products and deep oxidation products. For

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example, the olefin propylene can be directly oxidized with oxygen to form partial oxidation products, e.g., propylene oxide and acrolein, and deep oxidation products, e.g., carbon dioxide and water. (Fig. 1)

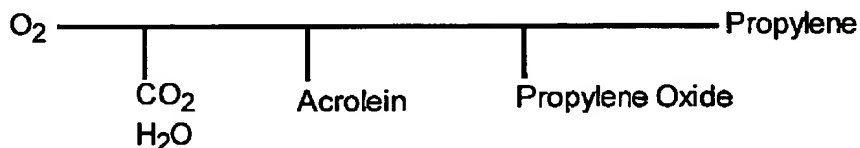


Fig. 1. (Direct Oxidation)

In contrast, the outcome of hydro-oxidation processes includes not only partial oxidation and deep oxidation products, but hydrogenation products from the reaction of organic substrate with hydrogen, as well as water from the direct combustion of hydrogen with oxygen ($H_2 + O_2 \rightarrow H_2O$). As an example, the hydro-oxidation of propylene with oxygen in the presence of hydrogen may result in the combustion of propylene with oxygen to partial oxidation products, e.g., propylene oxide and acrolein, and deep combustion products, e.g., carbon dioxide and water; the hydrogenation of propylene with hydrogen to form propane; and the combustion of hydrogen with oxygen to form water, as shown in Figure 2.

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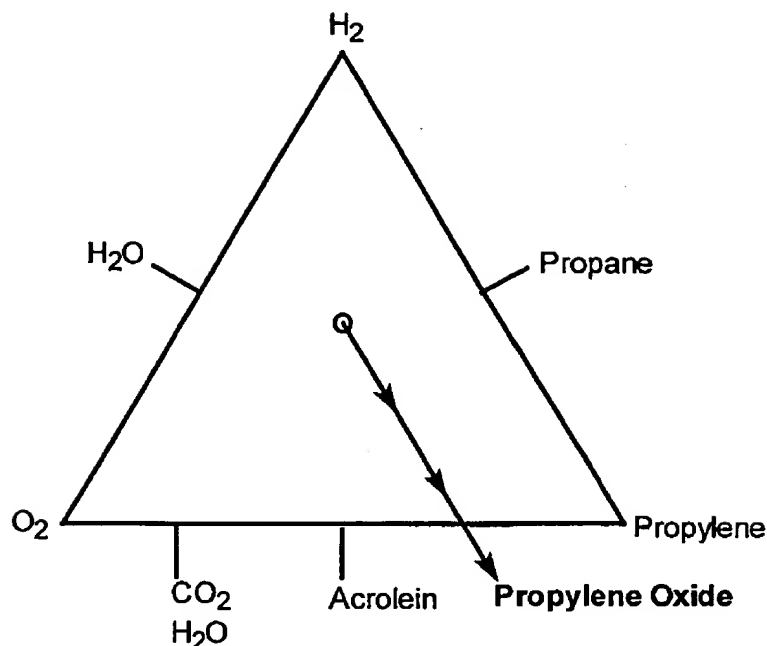


Fig 2. (Hydro-Oxidation)

(The bold arrow in Figure 2 represents the different pathway by which propylene oxide is formed when hydrogen is added to the oxidation process.) By comparing Figure 2 with Figure 1, it is seen that more reaction pathways are operable in hydro-oxidation processes than direct oxidation processes, therefore, hydro-oxidation processes require significantly different catalyst design and control over selectivities. Yet, a hydro-oxidation catalyst properly designed can provide for better selectivity to desired partial oxidation products, such as propylene oxide, particularly, when organic reactants having 3 or more carbon atoms are involved. Since Iwakura relates to the preparation of direct oxidation catalysts solely for a C₂ hydrocarbon reactant (ethylene), and since, in contrast, the claims relate to preparation of hydro-oxidation catalysts, preferably for C₃ or higher oxidations, Iwakura does not anticipate the claimed invention.

The Examiner argues that Applicants' limitation to intended use of the catalyst does not lend patentable weight to the claims. Applicants cannot agree. If the Examiner were correct, then no company would fund hydro-oxidation research, because prior art references drawn to generic techniques for preparing direct oxidation catalysts would anticipate and render obvious solutions

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to problems involving hydro-oxidation catalysts. Unfortunately, the catalysis art is so highly *unpredictable* that the prior art is not helpful, and the inventors' present employer and its competitors have been funding research for years on the preparation and development of hydro-oxidation catalysts for improved hydro-oxidation processes. (Witness the explosion of patent applications on hydro-oxidation chemistry since 1996; references to be provided upon request.) Applicants maintain the opinion that the intended use limitation to hydro-oxidation may be properly employed with other claim limitations to distance the claimed technology from the prior art of direct oxidation, thereby lending patentable weight to the claimed invention.

Regarding Claims 4-7 and 9, Iwakura discloses alcohol as a solvent for the amine complex. There is no recognition that the successful preparation of a hydro-oxidation catalyst requires a reducing agent, which may be satisfied by an alcohol.

Regarding Claim 37, Iwakura's disclosure of air oxidation does not embrace the claimed hydro-oxidation process requiring material quantities of both oxygen and hydrogen.

In view of the above, it is submitted that Iwakura does not anticipate amended Claims 1-7, 9, 18, 22-27, and 29-32. Accordingly, it is requested that the novelty rejection in view of Iwakura be withdrawn.

4. Concerning the Rejection of Claims 1, 4-5, 18, 22-23, 26-28, and 20-32

Claims 1, 4-5, 18, 22-23, 26-28, and 20-32 stand rejected as being anticipated by US 5,480,854 (hereinafter "Rajaram et al."), for the reasons of record. This rejection, as it might apply to the amended claims, is traversed for the following reasons.

Rajaram et al. teaches the preparation of a catalyst comprising noble metal particles, such as gold, uniformly incorporated into an oxide support, including titania (TiO₂), but preferably, ceria (CeO₂). The catalyst is prepared by *co-precipitation* from a solution containing precursor compounds of the metal

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oxide and the noble metal, or alternatively, a noble metal colloid containing small particles of noble metal suspended in a colloidal gel. The noble metal colloid is prepared by adding a reducing agent to a noble metal precursor compound. (Rajaram et al., Column 5, lines 63-67, onto Column 6, lines 1-5). See Example 3 of Rajaram et al. wherein solutions of cerium nitrate and chloroauric acid are mixed at controlled pH with NaOH to form a *precipitate*. See Example 8 of Rajaram et al. wherein a solution containing $ZrOCl_2$ and chloroauric acid, are treated with a reducing agent at controlled pH to *precipitate* gold particles into zirconia.

In contrast, Applicants' claims refer to impregnation of a gold compound onto a support, not precipitation or co-precipitation of such. Moreover, the instant claims require impregnation of a reducing agent; whereas the cited reference refers to a co-precipitation technique with a reducing agent. The differences between impregnation and precipitation or co-precipitation are well-documented in the art. Refer, for example, to the following citations, copies of which can be found in the file of the previous application:

"Either of two types of processes, generally termed the *precipitation method* and the *impregnation method*, is commonly used for making catalysts."

Heterogeneous Catalysis in Practice, Charles N. Satterfield, Professor of Chemical Engineering MIT, McGraw-Hill Book Company, New York, 1980, pp. 70.

Satterfield also teaches at page 70 that precipitation involves "in the initial stages the mixing of two or more solutions or suspensions of material, causing precipitation ". In contrast, referring to the impregnation method, Satterfield teaches:

"*Impregnation* is the easiest method of making a catalyst. A carrier, usually porous, is contacted with a solution, usually aqueous, of one of more suitable metallic compounds."

Heterogeneous Catalysis in Practice, Ibid., p. 71.

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Other references also recognize the differences between the impregnation and precipitation methods. See, for example, the following references, copies of which can be found in the previous application:

** Table XIII, wherein the methods of preparing a sulfur-free Ni/SiO₂ catalyst are disclosed as either:

"Various methods of coprecipitation at pH 8.5," or

"Dry mix," or

"Impregnation at 75°C."

"Structure and Activity of Silica-Supported Nickel Catalysts," by J. W. E. Coenen and B. G. Linsen, in *Physical and Chemical Aspects of Adsorbents and Catalysts*, B. G. Linsen, editor, Academic Press, New York, N.Y., 1970, p.501.

** "The active precursor can be applied onto the support by:

-adsorption

-impregnation and drying

-precipitation".

J. W. Geus, "Production and Thermal Pretreatment of Supported Catalysts," in *Preparation of Catalysts III*, G. Poncelet, P. Grange, and P. A. Jacobs, editors, Elsevier Science Publishers B.V., Amsterdam, 1983, p. 6.

For comparative purposes, Rajaram et al. also refers to impregnation. Specifically, Rajaram et al. teaches impregnation of a noble metal precursor compound onto a pre-formed metal oxide support with calcination (*high temperature heating*) for reduction to noble metal particles. See Rajaram et al. at Example 2 (Column 8, lines 57-62), Example 3 (Column 11, lines 49-50), and Comparative Example (Column 14, lines 60-63), wherein a Pt precursor is impregnated onto a CeO₂ support and then the support is calcined at 500°C to effect reduction to platinum particles. Nowhere is there mention or recognition of *impregnating a solution containing a reducing agent* onto the support. Moreover, the Au-CeO₂ impregnated catalyst, *absent impregnated reducing agent*, is shown in Table 5 (Column 12) to have absolutely no activity towards CO oxidation and NO_x reduction, thereby suggesting that impregnation is not a good method of preparing a gold oxidation catalyst.

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It is further noted that Rajaram et al. pertains to preparing catalysts intended for use in oxidizing carbon monoxide and hydrocarbons completely to carbon dioxide and water, while simultaneously reducing nitrogen oxides (NOx). Such catalysts are used in pollution control, in vehicle exhaust and internal combustion engines. In contrast, the instant claims pertain to preparing a catalyst for oxidation with oxygen in the presence of hydrogen. The objective as seen in the examples of the specification is to prepare an active oxidation catalyst for selective partial oxidation, not complete combustion (CO₂, H₂O) or NOx reduction. The use limitation clearly lends credibility to the patentability of the claims over Rajaram et al..

In view of the above, amended Claims 1, 4-5, 18, 22-23, 26-28, and 20-32 are not anticipated by Rajaram et al. It is therefore requested that the novelty rejection be withdrawn.

5. Concerning the Rejection of Claims 1-7, 9, 18, 22-23, 25-29, and 31-32

Claims 1-7, 9, 18, 22-23, 25-29, and 31-32 stand rejected as anticipated by US 4,937,219 (hereinafter "Haruta"), for the reasons of record. The Examiner especially points to Example 12 therein. This rejection, as it might apply to amended Claims 1-7, 9, 18, 22-23, 25-29, and 31-32, is traversed for the following reasons.

Haruta discloses a variety of *precipitation* methods for depositing gold onto an alkaline earth oxide or composite oxide with other metals, such as titanium, iron, cobalt, and nickel. The entire reference, *including Example 12*, embraces precipitation. In a first step, Example 12 teaches impregnation of magnesium nitrate onto a honeycomb support followed by calcination at high temperature to pre-form a support having MgO deposited thereon. *This first step merely pre-forms a support and has no relevance to the claimed impregnation of a gold compound and a reducing agent onto the support.* In a second step, the pre-formed honeycomb support is immersed in a solution of chloroauric acid; pH is adjusted to 10.2; and a reducing agent, magnesium citrate, is added dropwise slowly and with stirring for 1 hour. *This second step clearly embraces deposition-*

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precipitation by adding one solution (reducing agent solution) slowly at controlled pH to another solution (gold solution) to precipitate gold salts onto the support. This step does not involve impregnation, which would involve taking a solution of a gold compound and a reducing agent and adding it directly onto the solid support. Afterwards, Haruta teaches that the solution and the honeycomb support are separated, presumably by filtration. The resulting filtrate solution is adjusted to pH 12 and admixed with formalin to induce precipitation of any gold remaining in solution. This last step merely tests for gold remaining in solution, and as such, the step is irrelevant to Applicants' claims. In contrast, the instant claims pertain to impregnation of a gold compound and impregnation of a reducing agent onto a support. The art clearly documents and recognizes the distinctions between precipitation methods and impregnation methods. (Vide supra.)

Regarding Claim 9, Haruta at Example 12 employs water as a solvent, but makes no disclosure that the magnesium citrate reducing agent may also function as a solvent.

Regarding Claims 22-23 and 27-28, Haruta does not disclose impregnation, as explained hereinabove.

Accordingly, it is submitted that amended Claims 1-7, 9, 18, 22-23, 25-29, and 31-32 are not anticipated by Haruta. It is therefore requested that the novelty rejection be withdrawn.

6. Concerning US 6,255,499 (Kuperman et al.)

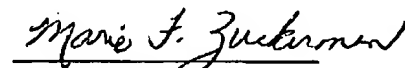
The Examiner refers to US 6,255,499 as pertinent for disclosing a process for making a composition comprising oxidized gold; however, the reference is not relied upon. Applicants note that US 6,255,499 was filed on April 7, 2000, claiming the benefit of priority provisional application 60/128,394, filed on April 8, 1999. It is observed that the instant application was also filed in the US on April 7, 2000, and claimed the benefit of a priority provisional

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application 60/128,390, which was also filed on April 8, 1999. Accordingly, the cited reference does not constitute prior art against the instant application.

In view of the arguments set forth hereinabove, it is submitted that in no manner whatsoever does Iwakura, Rajaram et al., or Haruta anticipate the claims. Amended Claims 1-32 and 37 are distinctly different from the cited references, and therefore meet all of the requirements for patentability. A Notice of Allowance is respectfully requested at the Examiner's earliest convenience.

Respectfully submitted,



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Attachment (Appendix A)